

Energy Straggling of Alpha Particles through Gases

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Measurements of the energy straggling of α particles through helium, air, argon, krypton, and xenon have been made for incident α energies between 2 and 4 MeV. These measurements were compared with the theoretical results of Bohr, Lewis, and Titeica. None of these theories gives satisfactory fits to the data, giving at best predictions within an order of magnitude of the experimental results. Capture and loss of electrons is a significant effect at these energies. The neglect of this effect and of the exact energy shell corrections, rather than an average shell correction, is believed to be the reason for the failure of these theories. Values of the stopping power for the α particles were also obtained. These were found to be in good agreement with proton energy-loss measurements after the usual corrections for mass and charge differences were made.

I. INTRODUCTION

SINCE the early work of Rutherford¹ and of Bragg,² the study of penetration of charged particles through matter has been important to nuclear physicists. Much of the stimulus for this work has come from the experimentalist's practical demands such as the need for energy corrections when particles pass through window foils, targets, etc. Another application has been the use of particle range or specific ionization as a means of energy measurements or of particle identification. Several reviews, both experimental and theoretical, have been published on the subject of atomic penetration.³⁻⁹

Heavy charged particles in their passage through matter lose energy almost entirely through inelastic collisions with the bound electrons in the atoms of the stopping material. This process is not a continuous one, but is made up of small but finite losses in a large number of collisions. It is to be expected that there will be statistical fluctuations in the total energy lost by particles with the same incident energy traveling through the same path length, and as a consequence there will also be fluctuations in the distance the incident particles will travel before being stopped. These fluctuation effects are known as "energy straggling" and "range straggling," respectively. As a measure of the straggling it is usual to specify either

the root-mean-square (rms) deviation Ω of the distribution about its mean value or its full width at half maximum η . Under the assumption that the straggling distribution is Gaussian these two quantities are related by $\eta = 2(2 \ln 2)^{1/2} \Omega$.

Although the theoretical understanding of both the energy loss and straggling is well advanced, the experimental information is rather incomplete. Measurements which have been made for incident particle velocities greater than those of the atomic electrons in the stopping material yield relatively good agreement with theory.⁴ For incident particle velocities comparable to that of the electrons, the process is complicated by the capture and loss of electrons by the incident ions. Measurements by Warsaw,¹⁰ Chilton *et al.*,¹¹ Hudson and Hofstadter,¹² Igo *et al.*,¹³ and Goldwasser *et al.*¹⁴ of energy loss and straggling show some discrepancy with theory.

Mason *et al.*¹⁵ have studied the straggling of 1-MeV protons in various gases. For small energy losses, the observed straggling was in fair agreement with the theories of Bohr^{16,17} and Lewis.¹⁸ A study of the energy straggling of α particles in metal foils has been made by Comfort *et al.*¹⁹ using 8.78 MeV ²¹⁰Po α particles. Large discrepancies with theory were found at low energies. The capture and loss of electrons by the incident ions was suggested as being primarily responsible for this discrepancy.

Most of the experiments on straggling^{11-14,16} have used metal foils as absorbers. In these measurements the observed straggling could have been due, in part,

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¹ E. Rutherford, *Phil. Mag.* **21**, 672 (1911).

² W. H. Bragg, *Studies in Radioactivity* (Macmillan and Company, Ltd., London, 1912).

³ U. Fano, *Ann. Rev. Nucl. Sci.* **13**, 1 (1963).

⁴ L. C. Northcliffe, *Ann. Rev. Nucl. Sci.* **13**, 67 (1963).

⁵ W. Whaling, in *Handbuch der Physik*, edited by S. Flugge (Springer-Verlag, Berlin, 1958), **34**, p. 193.

⁶ S. K. Allison and S. D. Warsaw, *Rev. Mod. Phys.* **25**, 779 (1953).

⁷ H. A. Bethe and J. Ashkin, in *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1953), Vol. I, part II, p. 166.

⁸ A. E. Taylor, *Rept. Progr. Phys.* **15**, 49 (1952).

⁹ H. K. Reynolds, D. N. F. Dunbar, W. A. Wenzel, and W. Whaling, *Phys. Rev.* **92**, 742 (1953).

¹⁰ S. D. Warsaw, *Phys. Rev.* **76**, 1759 (1949).

¹¹ A. B. Chilton, J. N. Cooper, and J. C. Harris, *Phys. Rev.* **93**, 413 (1954).

¹² A. Hudson and R. Hofstadter, *Phys. Rev.* **88**, 589 (1952).

¹³ G. I. Igo, D. D. Clark, and R. M. Eisenberg, *Phys. Rev.* **89**, 879 (1953).

¹⁴ E. L. Goldwasser, F. E. Mills, and A. O. Hanson, *Phys. Rev.* **88**, 1137 (1952).

¹⁵ D. L. Mason, R. M. Prior, and A. R. Quinton, *Nucl. Instr. Methods* **45**, 41 (1966).

¹⁶ N. Bohr, *Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd.* **18**, 8 (1948).

¹⁷ N. Bohr, *Phil. Mag.* **25**, 10 (1913).

¹⁸ H. W. Lewis, *Phys. Rev.* **85**, 20 (1952).

¹⁹ J. R. Comfort, J. F. Decker, E. T. Lynk, M. O. Scully, and A. R. Quinton, *Phys. Rev.* **150**, 249 (1966).

to the effects of nonuniformity of the foils.²⁰ Mason *et al.*¹⁵ have avoided this problem by using gases as the absorbers. Except for the latter reference no other measurements of energy straggling in gases have been published. In the present work we have measured the energy straggling of α particles in helium, air, argon, krypton, and xenon for incident α energies between 2 and 4 MeV. The results obtained are compared with theoretical predictions. Since values of the stopping powers for α particles could also be extracted from the experimental data these values are included with the experimental results.

II. EXPERIMENTAL PROCEDURE

The α -particle beam used in this experiment was provided by a 4-MV Van de Graaff accelerator. Beam currents of 0.5 to 1.0 μ A were used with an energy resolution of ± 2 keV obtained through a 90-deg momentum-analyzing magnet. A quadrupole magnet and a system of permanent and adjustable slits produce a well-defined beam on the first target of aluminum foil in the main scattering chamber (Fig. 1). The chamber was evacuated to less than 10^{-6} mm Hg by a 4-in. diffusion pump. The purpose of the first target was to provide a scattered beam reduced in intensity to within the working conditions of the detector and to strip the remaining electron from singly charged helium ions produced by the accelerator.

The apparatus used for the straggling measurements consists of a 26-cm long chamber with appropriate connections for the gas handling system. It was attached to the main chamber through a porthole located 45 deg from the incident beam direction. The chambers were separated by a thin window made from a polyvinyl chloride acetate copolymer. With a diameter of only 3 mm, the window also served as a collimator to define a secondary beam into the gas filled chamber. A Dubrovin mercury manometer was used to read the gas pressure; the gas temperature was taken to be the temperature of the chamber as measured with a thermometer attached to one of the outside walls.

After the beam had passed through the absorber, it was detected with a silicon surface-barrier detector. The pulses from the detector were passed through a charge-sensitive preamplifier and a low-noise biased amplifier and then recorded in a 128-channel pulse-height analyzer. The analyzer was calibrated, and its linearity was checked using a calibrated mercury-relay pulse generator.

Before any data were taken, the system was purged with the gas to be used in the subsequent series of runs. This was done by allowing the gas to leak slowly into the chamber while simultaneously pumping gas out of it. This method assured that there were no contaminants

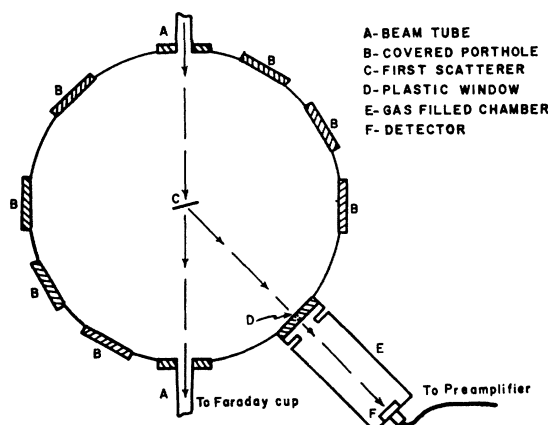


FIG. 1. Schematic diagram of experimental setup.

remaining in the system from the gases used previously. After purging the entire system, the pump was valved off and 20 mm Hg of the particular gas in use was introduced into the chamber.

A spectrum of the α particles, after passing through the gas, was taken with the multichannel pulse-height analyzer. No specific time period was used in taking the data. In order to obtain good statistics, the analyzer was allowed to run until the peak channel contained over 1000 counts. Spectra were taken in 0.25 MeV intervals for beam energies between 2 and 4 MeV. This procedure was followed each time for 20 mm Hg of helium, air, argon, krypton, and xenon. Spectra were also taken at the same incident energies with no gas absorber in the chamber.

From the spectra recorded it was possible to obtain the energy loss ΔE in the gas from the shift in the mean α pulse height and the energy straggling from the increase in the width of the pulse-height distribution.

III. RESULTS

A. Stopping Powers

Each spectrum obtained was plotted, and a smooth curve estimated by eye was drawn through the data points. Typical data to be analyzed are shown in Fig. 2. A resumé of the experimental data is given in Table I.

The stopping powers dE/dx at an energy E may be approximated by $\Delta E/\Delta x$ at E_{av} , where ΔE is the average energy loss of the α particles in passing through a thickness Δx of material, and E_{av} is the arithmetic mean of the incident and final energy. This procedure has been shown to be valid by Allison and Warshaw¹⁷ as long as ΔE is less than 20% of the incident energy E_0 . For losses greater than 20% of the incident energy, the values of $\Delta E/dx$ should be plotted at an effective energy

$$E_{\text{eff}} = E_{av} \{ 1 + [(\gamma - 1)/24](\Delta E^2/E_{av}^2) + \dots \},$$

where γ is the parameter obtained from the fitting of

²⁰ J. J. Ramirez and A. R. Quinton, Nucl. Instr. Methods 45, 353 (1966).

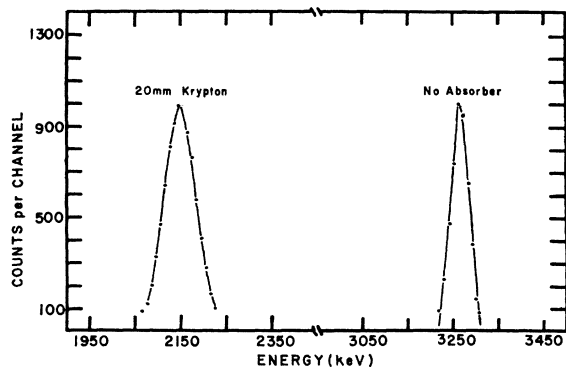


FIG. 2. Typical data to be analyzed.

the stopping power by a function of the form

$$-(dE/dx) = CE^{-\gamma}. \quad (1)$$

A least-squares fit of the experimental data to such a function gave values for γ as shown in Table II. These functions were later used in calculating the theoretical values of the straggling. The stopping powers $(-1/\rho)dE/dx$ are plotted in Fig. 3 as a function of the effective energy E_{eff} ; here ρ is the gas density. Also

TABLE I. Energy loss and straggling data.

Gas	Thickness (mg/cm ²)	Initial energy (MeV)	Final energy (MeV)	FWHM ^a (MeV)	Stopping cross sections (10 ⁻¹⁶ eV cm ²)
He	0.116	1.624	1.324	0.0416	17.2
	0.116	1.853	1.603	0.0446	14.3
	0.116	2.098	1.850	0.0397	14.2
	0.116	2.328	2.101	0.0417	13.1
	0.116	2.578	2.376	0.0370	11.6
	0.115	2.814	2.615	0.0407	11.5
	0.115	3.042	2.869	0.0374	10.0
	0.114	3.270	3.076	0.0370	11.3
Air	0.113	3.493	3.321	0.0362	10.1
	0.843	2.340	0.765	0.0776	89.9
	0.835	2.612	1.274	0.0743	77.1
	0.835	2.944	1.633	0.0670	75.6
	0.835	3.151	2.027	0.0600	64.8
Ar	0.835	3.418	2.370	0.0578	60.4
	1.19	1.624	0.379	0.0491	69.2
	1.19	1.853	0.649	0.0675	67.2
	1.19	2.098	1.018	0.0700	60.0
	1.18	2.328	1.355	0.0677	54.6
	1.17	2.578	1.674	0.0623	51.0
	1.17	2.814	1.964	0.0611	47.9
	1.17	3.042	2.242	0.0515	45.1
	1.17	3.270	2.498	0.0535	43.5
Kr	1.17	3.493	2.759	0.0514	41.4
	2.45	2.340	0.580	0.0696	99.8
	2.45	2.612	0.981	0.0768	92.5
	2.45	2.944	1.380	0.0817	88.7
	2.45	3.151	1.797	0.0944	76.8
Xe	2.45	3.418	2.083	0.0836	75.7
	4.07	2.612	0.357	0.0838	120.8
	4.07	2.944	0.696	0.1134	120.4
	4.07	3.151	1.126	0.1148	108.5
	4.07	3.418	1.536	0.1064	100.8

^a FWHM due to gas absorber only.

shown is the least-squares fit of the experimental points to the function $(-1/\rho)dE/dx = KE^{-\gamma}$. The other experimental points shown are the corresponding proton energy-loss data which have been calculated using the expression

$$-\frac{1}{\rho} \frac{dE}{dx} \Big|_{\alpha} = -\frac{1}{\rho} \frac{dE}{dx} \Big|_p \Big|_{z^*2}, \quad (2)$$

where z^* is the effective charge of the α particles as they penetrate the absorber. This effective charge was taken to be the charge of the incident ions, $z=2$.

B. Straggling

The spread due to straggling was obtained as follows: The spectrum of the α particles after passing through the absorber was plotted, and η , the full width at half maximum (FWHM), was determined; the FWHM was then determined for the spectrum of the α particles after passing through the evacuated chamber. The energy spreads were then subtracted in quadrature. This process eliminates the natural energy spread in the incident beam, the spread due to the electronics, and that due to the straggling through the first target and the plastic entrance window. Thus, $\eta^2 = (\eta_{\text{tot}})^2 - (\eta_{\text{instr}})^2$. These values obtained for η were plotted at the effective energy E_{eff} of the α particles in the stopping material as shown in Fig. 4. The experimental uncertainty was estimated to be $\pm 5\%$. The various theoretical predictions for the mean square spread Ω^2 were calculated for all gases and the values of the FWHM η given by $\eta = 2(2 \ln 2)^{1/2} \Omega$ were determined. These theoretical curves are also shown in Fig. 4.

IV. THEORY

Several theories on energy loss have been formulated. The most prominent ones are the classical-mechanical theory of Bohr^{16,17,21} and the quantum-mechanical theories of Bethe²² and Bloch.^{23,24} The Bloch theory is more comprehensive and contains the other two theories as limiting cases.

The energy-loss equations give the average energy loss suffered by a charged particle in traversing some

TABLE II. Values of K and γ obtained from a least-squares fit to the experimental values of the function $(-1/\rho)dE/dx$ (MeV cm²/mg) = $KE^{-\gamma}$, where E is given in MeV.

	K	γ
He	3.04	0.64
Air	2.32	0.61
Ar	1.05	0.47
Kr	0.82	0.44
Xe	0.65	0.33

²¹ E. Segrè, *Nuclei and Particles* (W. A. Benjamin, Inc., New York, 1965), p. 17.

²² H. Bethe, *Ann. Physik (Leipzig)* (5) 5, 325 (1930).

²³ F. Bloch, *Ann. Physik (Leipzig)* (5) 16, 285 (1933).

²⁴ F. Bloch, *Z. Physik* 81, 363 (1933).

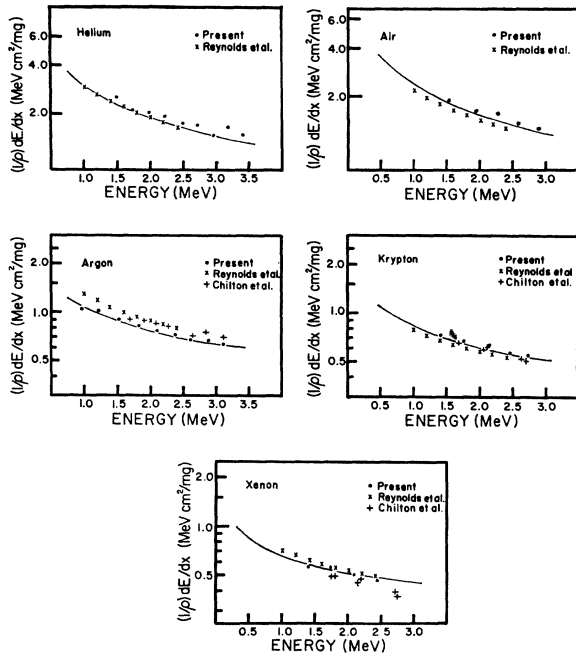


FIG. 3. α -particle stopping powers. The values of Reynolds *et al.* and Chilton *et al.* (Refs. 9 and 11) are proton measurements with the usual corrections for mass and charge differences. The solid curve is a least-squares fit to the present values.

stopping material. Any given particle loses energy in small but finite amounts, a large number of collisions being required to reduce its energy by an observable amount. As a result particles of the same initial energy which have all traveled the same path length show statistical fluctuations in the energy lost. If ΔE is the energy loss of a particular particle in passing through a given layer of material and $\langle \Delta E \rangle$ is the average value of the energy loss for a number of identical particles of the same incident energy, then the mean square fluctuation is defined as

$$\Omega^2 = (\Delta E - \langle \Delta E \rangle)^2. \quad (3)$$

The simplest of the straggling expressions is that of Bohr.^{17,18} Using the standard deviation of a Gaussian distribution of energies, he obtains

$$\Omega^2 = 4\pi e^4 z^2 N Z \Delta x, \quad (4)$$

where Δx is the thickness of the material through which the particle passes, ze is the charge of the incident particle, and N is the number of atoms per cm^3 of stopping material with atomic number Z . A plot of the predicted values of $\eta = 2(2 \ln 2)^{1/2} \Omega$ yields a straight line in this case since there is no energy dependence.

From Bethe's treatment²⁵ one finds that

$$\Omega^2 = 4\pi e^4 z^2 N \left(Z' + \sum_n \frac{I_n Z_n}{m v^2} \ln \frac{2m v^2}{I_n} \right) \Delta x. \quad (5)$$

²⁵ M. S. Livingston and H. Bethe, *Rev. Mod. Phys.* **9**, 245 (1937).

Here Z' is the total number of effective electrons, I_n is the average excitation energy of the Z_n electrons in the n th atomic shell, k_n is a constant taken to be $\frac{4}{3}$ for all electron orbits, m is the mass of an electron, and v is the velocity of the incident ion.

By applying the results of Bloch, Titeica²⁶ has been able to express the straggling width as

$$\Omega^2 = 4\pi e^4 z^2 N Z \left\{ 1 + \frac{4 E_{\text{kin}}}{3 m v^2} \left[\ln \frac{2m v^2}{I} + \psi(1) - \text{Re} \psi \left(1 + \frac{i z e^2}{\hbar v} \right) \right] \right\} \Delta x. \quad (6)$$

E_{kin} is the average kinetic energy per electron of the electrons in the stopping material, ψ is the logarithmic derivative of the Γ function, and $\text{Re} \psi$ denotes the real part of ψ .

The Titeica values were obtained by doing a numerical integration of Eq. (6) over the path length. This was necessary since the velocity of the α particles varies along the path traveled. The least-squares fits of the present data to Eq. (1) were used in calculating these velocities for the various gases. The values for the average excitation energy I were taken from the tables of Fano.³ These tables do not give a value of I for xenon. As is suggested by Bethe and Ashkin,⁷ the value $I = 9.2Z$ eV was used in this case. The average kinetic energy per electron of the electrons in the stopping material has been evaluated by Hund²⁷ from a Fermi-Thomas model of the atom. The expression $E_{\text{kin}} = 20.8Z^{4/3}$ eV was obtained. In making calculations for air, a mixture of 80% nitrogen and 20% oxygen was assumed, thus an average value $Z = 7.2$ was used. $\psi(1) = -\delta = 0.57722$ was used for the logarithmic derivative of the Γ function

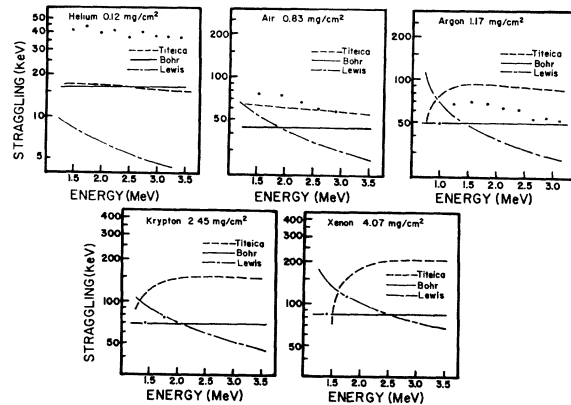


FIG. 4. Energy straggling of α particles. The experimental values are known to $\pm 5\%$.

²⁶ S. Titeica, *Bull. Soc. Roumaine Phys.* **38**, 81 (1939).

²⁷ F. Hund, in *Handbuch der Physik*, edited by H. Geiger and K. Scheel (Julius Springer-Verlag, Berlin, 1933), 2nd ed., **24** (1), p. 622.

of 1, while

$$\operatorname{Re}\psi(1+iy) = -\delta + y^2 \sum_{n=1}^{\infty} n^{-1}(n^2+y^2)^{-1} \quad (7)$$

gives²⁸ the real part of $\psi(1+iy)$.

Another approach to straggling is provided by Lewis¹⁸ who makes an estimate of the straggling by considering the collisions to be independent so that the number of collisions in a short distance Δx is distributed according to Poisson's Law. This expression gives the dispersion in energy produced by collisions in Δx which result in energy losses ΔE as

$$\Omega^2 = 4\pi e^4 z^2 N Z (M/m) (\Delta E \Delta x / E). \quad (8)$$

The Lewis values were also obtained by doing a numerical integration of Eq. (8) over the path traveled since the amount of energy lost ΔE varies along the path.

V. CONCLUSION

It can be seen from Fig. 3, where the present α -particle stopping powers are compared to the corresponding proton values obtained by Reynolds *et al.*,⁹ and Chilton *et al.*,¹¹ that the use of Eq. (2) relating the energy loss of protons to that of α particles is a good approximation and that the relationship becomes better at higher energies. The capture and loss of electrons by the incident ions lends itself as a good explanation of the discrepancies. This process becomes more pronounced at lower energies, and thus the use of $z=2$ for the effective charge z^* is less valid.

In contrast to the energy loss information, it is apparent from Fig. 4 that none of the theoretical predictions agree very well with the experimental straggling results. This is in agreement with the results of both Comfort *et al.*¹⁹ and Mason *et al.*¹⁵ It is interesting to note that helium is the only gas for which the experimental results are much larger than any one of the theoretical curves. The charge exchange phe-

nomenon again offers a possible explanation. Ions with different charges will lose energy at different rates. Due to the capture and loss process an α particle will spend some time in the singly charged state and some time in the doubly charged state while traversing the absorber. The time spent in one state or another will be different for different ions, and thus they will arrive at the detector with different energies; therefore, the net effect of the capture and loss process is to further broaden the distribution. It is reasonable to believe that charge exchange between α particles and the helium atoms will be relatively large since they have identical atomic excitation and ionization energies, thus causing an anomalous large amount of straggling.

None of the theories include the effects of the charge exchange process. It would be of much interest to do calculations involving an effective charge for the incident ions, but they are beyond the scope of this paper. Furthermore, it is apparent that only the Titeica curve has the correct energy dependence at low energies. This observed dependence is the opposite dependence found by Comfort *et al.*¹⁹ He observed an increase in the amount of straggling in metal foils at low energies. No simple explanation is available for this apparent inconsistency between these two results. One could possibly ascribe this to the difference in the nature of the targets used, but it is not obvious why this should be so. The experimental techniques do differ markedly, however, with the present work giving a more direct method of obtaining straggling measurements, as a function of energy, for a given target thickness. Because of the method used and since the present results exhibit the same dependence of the Titeica curve which is based on Bloch's^{23,24} quantum-mechanical theory, we believe that the presently observed low-energy dependence is correct. It is believed that by doing a more exact calculation of the Titeica expression for straggling, including more exact values of the various terms that include the shell effects, it will be possible to obtain a much better agreement between theory and experiment. The Bohr theory may be used to compute the amount of straggling involved when only a rough estimate is required.

²⁸ M. Abramowitz and I. A. Stegun, in *Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables* (U. S. Government Printing Office, Washington, D. C., 1965), p. 259.